AUTOMATED QA/QC CHECK

FOR β-γ COINCIDENCE DETECTOR

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ABSTRACT

The Automated Radioxenon Sampler/Analyzer (ARSA) (Hayes 1999), built by Pacific Northwest National Laboratory (PNNL), can collect and detect several radioxenon isotopes. The ARSA is very sensitive to ^{133}Xe, ^{131m}Xe, ^{133m}Xe, and ^{135}Xe due to the compact high-efficiency β–γ coincidence detector it uses. As detection technology improves and more emphasis is placed on system automation, QA/QC checks become of increasing importance. By automating the QA/QC checks, the reliability of the data sets is ensured without the necessity of monitoring. Often, the automated QA/QC checks will be done on site, where radio-xenon gas is not readily available so it is important to be able to use sealed point sources as an alternative. Immediately after the initial calibration of the detector, a ^{137}Cs source, which has a 661.7-keV γ-ray, is used to generate both γ and β spectra. The two spectra are saved to file, are called the template spectrum, and will be used as the template to perform on site QA/QC checks.

The QA/QC check will automatically find the start and end points of the β spectrum and compare them with those from the template spectrum. A similar method is used for the γ spectrum; however, in a γ spectrum the 661.7-keV peak can be used for the high energy end. If the points do not match, a gain and offset can be calculated to make the QA/QC spectra match the template spectra.

A second and more advanced method is to take a β–γ coincidence spectrum for the template spectrum. A comparison of the QA/QC β–γ coincidence spectrum to the template spectrum will be used to determine what if any adjustments need to be made on the gain and offset. A detailed discussion of the QA/QC checks will be included in the paper.

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OBJECTIVES

Monitoring radioactive releases from nuclear explosions is a major component of the International Monitoring System (IMS) network (Hayes 1999). As part of the international effort to develop monitoring equipment, PNNL deployed the ARSA to Germany for an international demonstration of radionuclide equipment (Bowyer 1999). The ARSA measures radioxenon isotopes directly by first collecting and processing whole air samples that potentially contain a small amount of radioxenon (Bowyer 1999). After the samples are collected and processed and the xenon is isolated, it is counted in a nuclear detector. A critical component in the quantification of the radioxenon isotopes is the nuclear detector calibration. There are three major components to characterizing the ARSA nuclear detector: energy calibration, detection efficiency measurements, and spectral interference ratios. The nuclear detector is calibrated at the laboratory, but also requires monitoring in the field to ensure proper performance [to attain the lowest minimal detectable concentration (MDC) (McIntyre 2006)].

Figure 1. A: An ARSA type β-γ detector. The NaI(Tl) detector component is to the left and the gas-cell that slides into the NaI(Tl) is to the right. B: A Quad β-γ detector. The full detector is rendered on the left side and is comprised of 4 well-type CsI(Na) detectors and four gas-cells (β-cell). A magnified view of a single β-cell is given on the right.

Two similar styles of β-γ coincidence detectors are shown in figure 2. The detector on the left is the original ARSA detector and one on the right is an updated version called the Quad β-γ coincidence detector (Bowyer 1999 and Cooper 2005). Both nuclear detector types utilize an inorganic scintillator to detect x-rays and gammas and a gas-cell made of plastic scintillator to hold the gas sample and to detect outgoing charge particles, β, α, and conversion electrons (CE). In addition, both detectors are comprised of four gas-cells which individually measure one gas sample at a time. For the purposes of this paper the Quad detector was used to perform all test and measurements. The remainder of the paper will step through how The Quad β-γ coincidence detector is characterized and what steps can be taken to verify good performance while in the field.

RESEARCH ACCOMPLISHED

Laboratory calibration of a single β-γ detector involves use of ^{137}Cs as an initial check, followed by spiking the detector with radioactive gasses for precise determination of the nuclear detector characteristics. The three radioactive gases that PNNL uses are ^{133}Xe, ^{131m}Xe, and ^{222}Rn. Where possible, pure spikes are used; however, some contaminants are likely (e.g., ^{131m}Xe contaminant in a ^{133}Xe spike). These contributions are subtracted out during the calibration process to yield the correct parameters. All three spikes emit combinations of γ-rays and x-rays in coincidence with CE and β particles (see Table 1). Due to minor fluctuations in the gas processing system of the ARSA, ^{222}Rn gas can be introduced into the gas cell, along with the radioactive xenon isotopes. While this radon decays via alpha decay and is only detected in the beta spectrum, two of its subsequent daughter products, ^{214}Pb and ^{214}Bi, decay via β-γ coincidence. The dominant ^{214}Pb is shown in Table 1 as it is the primary decay product of ^{222}Rn that interferes with the radioxenon β-γ coincidence spectra.

Table 1. Energy and branching ratios for the three radioactive gas spikes.

	^{131m}Xe	^{133}Xe	^{214}Pb[a]
Half-life (days)	11.84	5.24	0.0186
Primary γ-ray Energy (keV)	163.9	81.0	351.87
γ-ray Abundance (%)	1.96	37	37.1
Primary X-ray Energy (keV)	30.0	31.0	77.1
X-ray Abundance (%)	54	48.9	11.0
β-particle End-point Energy (keV)		346	1023
β-particle Abundance (%)		100	100
Primary CE Energy (keV)	129	45	37
CE Abundance (%)	60.7	54.1	10.7

[a] ^{222}Rn Daughter

Detector Spike

The nuclear detector is first setup using a ^{137}Cs source to adjust the high voltage (HV) on both the β and the γ detectors using γ-singles to adjust the HV for the γ detector and β-γ coincidence to adjust the HV for the β detector. The high voltage adjustments set the range of energies displayed in the gamma and beta singles spectrum as well as the β-γ coincidence plane. Subsequent measurements using the radioactive gases calibrate the channel to energy scale for these spectra more accurately. As soon as the HV is set, a 3-day detector background is taken. The data collected during this background count, and for spike tests, includes three spectrum, γ-singles, β-singles and β-γ coincidence data. After all four well detectors have collected the background spectra, a series of spikes is put into each β-cell. The spikes are large enough (~30 Bq for ^{131m}Xe) to collect high statistics in a 1-hour count and begin with Rn. The second spike is ^{133}Xe and the final spike is ^{131m}Xe. After each set of data is collected, a pump and flush process is used for several days to remove the residual gas that adheres and penetrates the walls of the gas cell. Once the pump and flush cycle is complete the next set of radioactive gas is injected into the detector.

Detector Energy Calibration

The γ- and x-ray spectrum generated (Figure 2) from the radon daughters is used to calibrate the CsI(Na) or NaI(Tl) well detectors. This spectrum gives seven peaks (53.2 keV (γ-ray ^{214}Pb), 78.8 keV (x-rays ^{214}Pb), 241.9 keV (γ-ray ^{214}Pb), 295.1 keV (γ-ray ^{214}Pb), 351.9 keV (γ-ray ^{214}Pb), 609.3 keV (γ-ray ^{214}Bi), and 30 keV (CsI x-ray)). A linear fit to a plot of channel vs. energy gives the γ-ray energy calibration for the system.

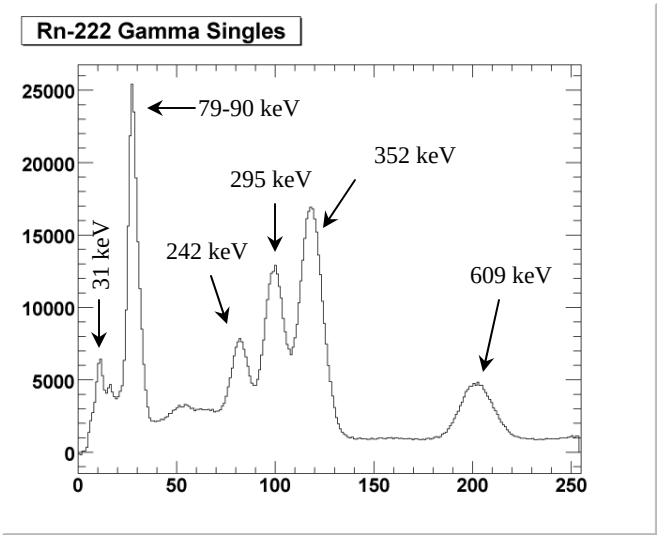

Figure 2. γ-singles spectrum from a ^{222}Rn spike. The peaks are primarily from the radon daughter ^{214}Pb with the highest peak being the 609-keV γ-ray from ^{214}Bi.

The CE and β end-point energies are used to calibrate the β-detector and entails using all three radioactive gas data sets. The ^{131m}Xe β-γ data give a peak at 129 keV for the β and 30 keV for the γ, which can be seen as a contaminant for ^{133}Xe in Figure 3. In Figure 3, the ^{131m}Xe peak is at roughly channel 32 on the abscissa. The remainder of the β energy calibration is done using end-point energies. To determine the end-point energy a region of interest (ROI) is made around a β-γ line. The ROI is then projected onto the x-axis to get a β spectrum. The channel at which the β continuum approaches zero is the end-point energy value.

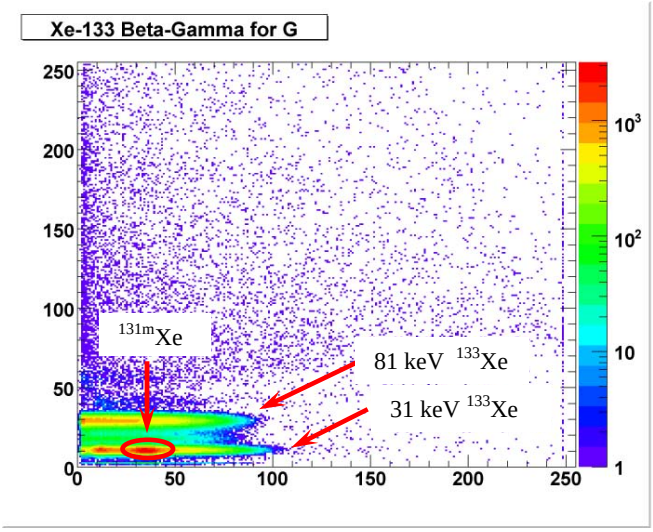

Figure 3. A β-γ spectrum from a ^{133}Xe spike with some ^{131m}Xe contamination.

Detector Efficiency

The β-γ efficiency measurement uses the same three data sets as are used to calibrate the β and γ energies: ^{131m}Xe, ^{133}Xe, and ^{214}Pb (^{222}Rn daughter). Due to the ~100% β and CE efficiencies, in general the number of counts in a β-singles spectrum, weighted by the branching ratio (BR) and efficiency (ε), will equal the number of counts in the

β-γ spectrum, weighted by its branching ratio and efficiency as shown in Equation 1. The BR is the percentage of particles, which decay by a particular decay mode with respect to the total number decays.

$$\frac{Counts_{\beta}}{Br_{\beta} * \varepsilon_{\beta}} = \frac{Counts_{\beta\gamma}}{Br_{\beta\gamma} * \varepsilon_{\beta\gamma}} \quad \textbf{(1)}$$

where,

$$\varepsilon_{\beta\gamma} = \varepsilon_{\gamma} * \varepsilon_{\beta} \quad \textbf{(2)}$$

And, in general, $Br_{\beta\gamma}$ equals Br_{γ} since there is an exact coincidence between the β and γ decay modes. In the case of ^{131m}Xe ε_{β} is assumed to be ~100% due to the 99.9% solid angle coverage for β's decays within the β-cell and the 129-keV CE that is a single line and not a beta continuum.

$$\varepsilon_{\beta\gamma} = \left(\frac{Counts_{\beta\gamma}}{Counts_{\beta}}\right)\left(\frac{Br_{\beta}}{Br_{\beta\gamma}}\right)\underbrace{\varepsilon_{\beta}}_{=1} \quad \textbf{(3)}$$

Since ^{133}Xe has a 31-keV x-ray in coincidence with a β continuum plus a 45-keV CE, it is possible to use the ^{131m}Xe γ efficiency to determine the ^{133}Xe-80 keV γ efficiency.

$$\varepsilon_{\gamma}^{(80keV)} = \left(\frac{Counts_{\gamma}^{Xe-133(80keV)}}{Counts_{\gamma}^{Xe-133(30keV)}}\right)\left(\frac{Br_{\gamma}^{Xe-133(30keV)}}{Br_{\gamma}^{Xe-133(80keV)}}\right)\varepsilon_{\gamma}^{(30keV)} \quad \textbf{(4)}$$

Equation 4 is possible due to the overlap in energies between ^{131m}Xe and ^{133}Xe at 30 keV (the detector will have the same γ efficiency for all γ-rays that occur at the same energy). By using the β–γ response from ^{214}Pb the γ efficiency at 80 keV is applied to determine the γ efficiency at 242, 295, and 352 keV. Once again this is due to overlapping γ energy, which in this case is at 80 keV.

$$\varepsilon_{\gamma}^{(X)} = \left(\frac{Counts_{\gamma}^{Pb-214(X)}}{Counts_{\gamma}^{Pb-214(80keV)}}\right)\left(\frac{Br_{\gamma}^{Pb-214(80keV)}}{Br_{\gamma}^{Pb-214(X)}}\right)\varepsilon_{\gamma}^{(80keV)} \quad \textbf{(5)}$$

Where, "X" can be 242, 295, or 352 keV.

As mentioned earlier, one complicating factor is any contaminants present in the spike. Figure 4 shows a ^{131m}Xe spectrum containing ^{133}Xe and ^{222}Rn contaminants. One method to remove the contaminants from the spectrum is to have a pure spectrum of ^{133}Xe and a pure spectrum of ^{222}Rn and subtract them from the ^{131m}Xe spectrum.

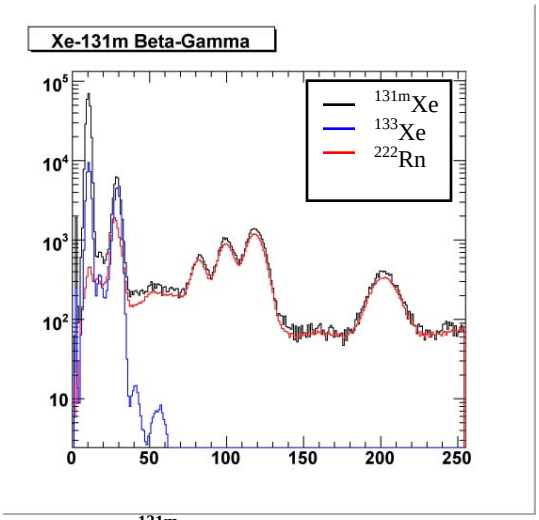

Figure 4. A ^{131m}Xe spectrum (black line) with both ^{133}Xe and ^{214}Pb contaminants present. The red spectrum is a pure ^{214}Pb spectrum that was normalized to the ^{131m}Xe contaminant level above 80 keV. Similarly, the blue spectrum is a pure ^{133}Xe spectrum also normalized to the ^{131m}Xe contaminant level above 30 keV.

The easiest way to do the subtraction is to first normalize the ^{222}Rn spectrum to the level of contaminant present using the 242-, 295-, and 352-keV peaks. After the spectrum is normalized, the ^{222}Rn is then subtracted from the ^{131m}Xe spectrum; this must be done before the ^{133}Xe contaminant level can be determined because ^{222}Rn contains an 80 keV peak from ^{214}Pb, which interferes with the 81-keV ^{133}Xe peak. After the ^{222}Rn is subtracted, the 80-0keV peak in the ^{131m}Xe spectrum should be solely produced by the ^{133}Xe contaminant, so normalization of the ^{133}Xe spectrum follows. The normalized ^{133}Xe (blue) and normalized ^{222}Rn (red) spectra are displayed in Figure 5.

β-γ Detector Auto-Calibration

As stated earlier it is important to have a well-calibrated detector during field operation. To accomplish this, an automated process has been developed that can track and adjust the energy calibration. ^{137}Cs is an easily obtained source that has a 661.7-keV γ-ray that will Compton scatter into the β detector. Figure 5 shows a typical β-γ spectrum taken using a ^{137}Cs source. The diagonal line in the spectrum is the constant energy line, where the β energy plus the γ energy is equal to 661.7 keV. The constant energy line ends well short of zero E_γ due to the maximum energies imparted to the Compton scattered electron, (reference Reeder 2004 describes the process in detail)

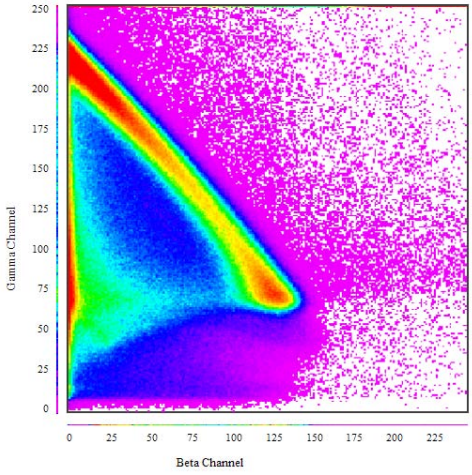

Figure 5. A β-γ spectrum taken using a ^{137}Cs source.

A method to verify and recalibrate a system in the field uses the β- and γ-singles spectra from a ^{137}Cs source to adjust gains and HV settings. The first step is to collect high-statistics β- and γ- singles from a fully calibrated detector, which will be referred to as a template spectrum (Figure 6). The template spectra will then be used as a comparison for what a calibrated detector should look like. When a detector is deployed, the first step is to acquire a β-singles and a γ-singles ^{137}Cs spectrum and compare the singles data with the template spectra. The comparison is done in two steps.

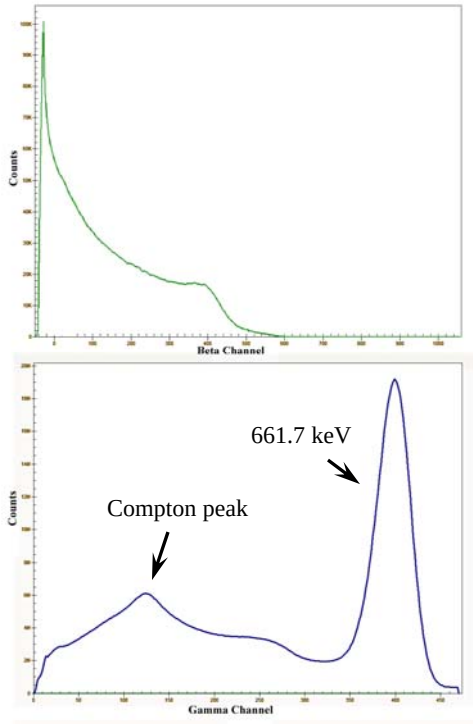

Figure 6. The top figure is a β-singles spectrum from Compton scattering using a ^{137}Cs source. The bottom spectrum is a γ-singles spectrum using a ^{137}Cs source.

First, the γ singles are analyzed to find the 661.7-keV peak and the Compton peak. The separation between the peaks represents the gain on the γ channel, which is then adjusted to match the template spectrum peak separation. Next, the 661.7-keV peak position is adjusted to the peak position in the template spectrum, giving the offset on the γ channel. Therefore, by adjusting the gain and offset the template spectrum, the γ spectrum and the calibration file should be aligned.

The second half of the processes is used to calibrate the β singles spectrum. This method finds the beginning and end-point of the spectrum. Using

$$\frac{\sqrt{\sum_{0}^{n} counts}}{\sum_{0}^{n} counts} = C, \qquad (6)$$

where C is 0.1 for finding the end-point. Similarly,

$$\frac{\sqrt{\sum_{n}^{250} counts}}{\sum_{n}^{250} counts} = C, \qquad (7)$$

where C is 0.05 for finding the beginning point. In both cases, *n* is the channel number. The value of C is determined empirically to give the best statistical results. The two points, beginning and end-point, are compared with those calculated from the β-template spectrum to determine the gain and offset for the β-cell.

CONCLUSIONS AND RECOMMENDATIONS

All detectors must be fully characterized before use, and this is especially important for fielded systems. For many detectors and for field calibration verification, this is a routine process using sealed sources. However, for detectors used in the ARSA and similar coincidence counting systems, sealed sources do not make the best choice for the initial energy calibration. Instead the use of radioactive gases gives a more accurate method of determining energy calibrations and interference ratios.

The initial characterization of a β-γ detector involves injecting multiple sources to cover the complete spectrum of energies. The three gases used are ^{131m}Xe, ^{133}Xe, and ^{222}Rn, which must be pure and high activity (30-60 Bq) to obtain high enough statistics to minimize the errors for a 1-hour acquisition. By carefully analyzing each data set the γ-ray efficiency, β-particle efficiency, γ-ray energy calibration, and β-particle energy calibration are determined.

Since a detector is only as good as its calibration the next step is to maintain the calibration of the detector system. This is most easily done in an automated fashion using a ^{137}Cs source. Through comparing new calibration files to template spectra it is possible to adjust the gain and offset to regain a good energy calibration on a detector system that otherwise would give misleading information.

Further development of the auto-calibration is in order. Some potential changes involve using a chi squared minimization of a β-γ calibration file with a β-γ template spectrum. Another direction involves using a comparison of principal components to determine the line of constant energy in a β-γ spectrum.

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